#### **Short communication**

# Mercury in bottom sediments of Russian Arctic lakes



# Tatsiy Yu.G.\*

Vernadsky Institute of Geochemistry and Analytical Chemistry of the Russian Academy of Sciences, Kosygin St., 19, Moscow, 119991, Russia

**ABSTRACT.** This article presents the results of studying the distribution of mercury in the cores of bottom sediments of both background and technogenic Arctic lakes in three regions: the Kola North, Malozemelskaya tundra and northwest Siberia. For all cores, the elemental composition, granulometry, and loss on ignition (LOI) were determined layer by layer. Mercury showed a significant excess of the local background in the near-surface sedimentary layers of most lakes. Fluxes of mercury from the pre-industrial period to the present were calculated. We discuss the possibility of technogenic pollution due to transboundary transfer.

Keywords: Arctic lakes, bottom sediments, mercury, sedimentation rate

# **1. Introduction**

Studies of dated cores from Arctic lakes have been carried out since the early 1990s to determine the fluxes and history of heavy metal sedimentation. This is largely driven by the need to understand the spatial and temporal pollution trends in the Arctic and the impact pathways for wildlife and humans. Arctic lakes are sensitive indicators of global environmental and climate change as well as of the impact of regional and transboundary transport of pollutants. A huge number of lakes are concentrated in the Arctic regions of the continental tundra. In the Russian part of the Arctic, there are over 2.5 million of them. However, only about 19 thousand lakes have an area of more than 1 km<sup>2</sup>. Nevertheless, the total area of the entire water surface of the Arctic lakes reaches 160,000 km<sup>2</sup>, which is slightly less than half the area of all natural water bodies in Russia.

Mercury in the Arctic is a subject of particular interest. Mercury, as a global pollutant, is a cause for concern due to its potential toxicity and increased bioaccumulation in the Arctic food chain, including humans.

In recent decades, significant advances have been made in describing the cycle and behavior of mercury in freshwater environments. A large amount of new data on Hg concentrations, forms, and fluxes has been provided and summarized for water and sediments, but all this applies mainly to the Canadian Arctic and Alaska (Chételat et al., 2015; Douglas et al., 2012). Only a few

\*Corresponding author.

E-mail address: yutatsy@mail.ru (Yu.G. Tatsiy)

*Received:* June 16, 2022; *Accepted:* July 19, 2022; *Available online:* July 31, 2022

studies were devoted to the behavior of mercury in lake sediments in the Russian Arctic (for example, Dauvalter and Kashulin, 2018; Tatsii and Baranov, 2022).

Biogeochemical processes that control the accumulation of mercury in sediments are not well understood. Additionally, the Arctic is undergoing profound environmental changes associated with climate warming, and preliminary evidence suggests that this may affect the cycle and bioaccumulation of mercury.

The aim of the work was to study the distribution of mercury in lake sediment cores from various regions of the Russian Arctic, both background and subject to technogenic influence, to assess the dynamics of mercury sedimentation and to identify the anthropogenic contribution.

#### 2. Materials and methods 2.1. Study area and sample collection

Samples of bottom sediments were taken in several regions:

- in the lakes of northwest Siberia (Langtibeito Lake and Goltsovoye Lake) by colleagues from Tyumen State University in 2011;
- in the lakes of the Malozemelskaya tundra by the British expedition in 2017, and
- in the Kola North by the author and colleagues from Vernadsky Institute of Geochemistry in 2018.

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Lake	Coordinates		Area, km <sup>2</sup>		Height	Average	pН	Core
	Latitude	Longitude	Lake	Watershed	a.s.l., m	depth, m		lengths, cm
Langtibeito	71.063917	70.321806	10.3	8.2	43.4	3.0	7.71	57 and 10
Goltsovoye	71.423333	78.849444	3.87	8.1	9.6	3.5	7.39	48 and 23
NARY 1_2	68.53787	53.71337	1.5	-		0.6	5.9	8
NARY 4_2	68.35489	53.96419	0.07	-		0.7	7.3	9
NARY 9_1	68.3535	53.9571	0.04	-		1.1	8.6	10
Dolgoe	69.454966	31.902271	0.56	12.7	210.0		6.53	25
Keinojärvi	69.45152778	30.65202778	0.14	4.75	131.1		7.12	40

Table. Basic data of the studied lakes

All lakes are located in the Arctic tundra subzone that is characterized by a long duration of snow cover (about 10 months) and a short summer with low temperatures  $(1-5^{\circ}C)$ . The main source of income for tundra lakes is atmospheric precipitation and water formed during the seasonal thawing of permafrost. All lakes, except for the Kola lakes, are thermokarst.

For the sampling of the bottom sediments, different types of core samplers with tubes of different diameters were used. Under field conditions, all cores were cut into slices 1 and 0.5 cm thick.

Lakes vary in both size and position. Langtibeito and Goltsovoye are located far from the sea coast; NARY 1-2 is located about 5 km from the coast, and NARY 4-2 and NARY 9-1 are located on Lovetsky Island at a distance of 300 m from each other and about 1.5 km off the coast. The Kola lakes are located far from the coast and are subject to technogenic influence, as they are located in the zone of influence of large metallurgical smelter.

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#### 2.2. Core dating and mercury determination

All cores were radiometrically dated by measuring the activity of isotopes <sup>210</sup>Pb and <sup>137</sup>Cs. Loss on ignition (LOI) was determined by keeping in a furnace at 550°C for at least 2 hours (to constant weight). For individual layers, granulometric analysis by dry sieving for five fractions was performed.

Mercury was determined in all samples from a sample of 20 to 100 mg (n=3) by pyrolysis of a solid sample with a preliminary accumulation on a gold collector and recording on an AA spectrometer. Calibration was carried out by saturated mercury vapor. The elemental analysis (63 elements) after acid decomposition was performed using ICP-MS at Vernadsky Institute of Geochemistry.

#### **3. Results and discussion**

Based on the dating results, we estimated sedimentation rates, background concentrations and enrichment factors relative to the pre-industrial period (before  $\sim 1850$ ). The enrichment factors were calculated relative to the local background levels of mercury (and some other elements), which were determined as the average value of those measured in several lower slices of the cores.

Mercury background levels were 5.2, 4.3, and 7.0 ng/g for lakes Goltsovoye, Langtibeito, and NARY 1-2, respectively. For lakes subjected to technogenic influence, Dolgoe and Keinojärvi, background concentrations were 103 and 78 ng/g, respectively. The higher background levels of mercury in these lakes appear to be related to the underlying geology.

The concentration profiles of Hg in the bottom sediments themselves only indirectly reflected the nature of the metal entering the sediments and its source. The flows of elements in the sediments are more informational and complementary. The overall sedimentation rates (black curves in the figure) in all lakes, except for Langtibeito, gradually decrease to the surface layers, while Hg concentrations increase. The average mercury fluxes calculated from deposition rate, slice volume and weight, and mercury concentration in each layer (red curves) show gradual increases in the Langtibaito and Goltsovoe cores and decreases in all NARY cores.

The background fluxes of the total Hg (before 1850) in the studied background lakes were similar and as accounted for 5.1, 2.5, and 1.6  $\mu$ g/m<sup>2</sup>year for Goltsovoe, Langtibeito, and NARY 1-2, respectively. Compared to the preindustrial period, the Hg flux had a 3.9-fold increasein Goltsovoye and a 7.9-fold one in NARY 1-2. In Langtibeito, there was a 16.4-fold increase, which is unusually large for polar lakes and may indicate a possible strong anthropogenic influence. In technogenic lakes, the background fluxes were significantly higher: 11.2 µg/m<sup>2</sup>year in Dolgoe and 9.9 µg/m<sup>2</sup>year in Keinojärvi. In Keinojärvi, this flow increased by a factor of 5, while in Dolgoe it did not change. It should be noted that the present values of Hg fluxes exceed the atmospheric deposition estimated for the Canadian Arctic as 6-12  $\mu$ g/m<sup>2</sup> per year (Chételat et al., 2015).



**Fig.** Features of the Hg distribution in Arctic lakes. Top – concentration distribution of Hg and dating; bottom – curves of deposition flux (black), mercury fluxes (red), and mercury fluxes corrected for Sc and Fe (blue).

To identify the conditionally "anthropogenic" flux in background lakes, the formula proposed by Norton and Kahl (1987) was used to isolate anthropogenic concentrations. We used it to isolate Hg fluxes of non-lithogenic origin. The results are reflected in the blue curve at the bottom of the figure. Fe, Ti, Mg, and Sc were used as reference elements, which gave almost identical curves. The shaded region can be represented as a "conditionally anthropogenic" Hg flux, i.e. non-lithogenic. This flux, in addition to direct atmospheric deposition, may include watershed inputs as well as those from permafrost thawing.

The change in conditionally lithogenic fluxes according to the core data from the studied lakes ranged from 3 to 7  $\mu$ g/m<sup>2</sup> year during the post-industrial period. The insignificance of the lithogenic contribution was also reported by Lindqvist et al. (1991) and Swain et al. (1992).

In Langtibaito, the total Hg influx increased approximately since the 1940s, reaching 41.5  $\mu$ g/m<sup>2</sup>year now, which is rather high for an Arctic thermokarst lake. Two episodic increases around the early 1960s and late 1970s were not accompanied by changes in sedimentation rate but correlated with LOI and particle size distribution.

In Goltsovoe Lake, the anthropogenic Hg flux began to gradually increase approximately since 1970, with an episodic sharp increase around 1980. Sharp increases in Hg flux in both lakes occurred approximately at the same time (late 1970s and early 1980s).

In NARY 1-2, some increase in Hg flux occurred approximately in the same period, around the 1980s. However, all this took place with a gradual decrease in deposition, which became minimal in the upper layer. At the same time, the concentration in the upper layer reached its maximum values.

For NARY 4-2 and 9-1, background fluxes, as well as adjusted fluxes, could not be determined because the depth of the cores did not reach the pre-industrial levels.

## 4. Conclusions

A layer-by-layer analysis of two cores of bottom sediments showed that the Hg distribution differs significantly from the distribution of other elements by much more intensive enrichment of surface layers.

Studies have revealed that the concentration distribution along the cores does not always unambiguously characterize the entry of mercury into the bottom sediments of Arctic lakes. Analysis of Hg fluxes indicates a non-lithogenic mercury flux. The problem of identifying a purely anthropogenic flux remains unresolved.

Based on the results of the distribution of elements in sediments, all lakes for all studied elements (except for Hg) are background. For Hg, the lakes are not background but remain unpolluted.

## Acknowledgements

The research was supported by the RSF grant 22-17-00061

## **Conflict of interest**

The authors declare no conflicts of interest.

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